# Generation of higher order nonclassical states via interaction of intense electromagnetic field with third order nonlinear medium

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#### Abstract

Interaction of intense laser beam with an inversion symmetric third order nonlinear medium is modeled as a quartic anharmonic oscillator. A first order operator solution of the model Hamiltonian is used to study the possibilities of generation of higher order non-classical states. It is found that the higher order squeezed and higher order antibunched states can be produced by this interaction. It is also shown that the higher order nonclassical states may appear separately, i.e. a higher order antibunched state is not essentially higher order squeezed state and vice versa.

#### 1 Introduction

A nonclassical state of the electromagnetic field is one for which the Glauber-Sudarshan P-function [1] is not as well defined as the probability density is [2]. To be precise, if P-function becomes either negative or more singular than delta function, then we obtain a nonclassical state. The nonclassical states do not have any classical analogue. Commonly, standard deviation of an observable is considered to be the most natural measure of quantum fluctuation [3] associated with that observable and the reduction of quantum fluctuation below the coherent state level corresponds to a nonclassical state. For example, an electromagnetic field is said to be electrically squeezed field if uncertainties in the quadrature phase observable X reduces below the coherent state level (i.e.  $(\Delta X)^2 < \frac{1}{2}$ ) and antibunching is defined as a phenomenon in which the fluctuations in photon number reduces below the Poisson level (i.e.  $(\Delta N)^2 < \langle N \rangle$ ) [4, 5]. Standard deviations can also be combined to form some complex measures of nonclassicality, which may increase with the increasing nonclassicality. As an example, we can note that the total noise of a quantum state which, is a measure of the total fluctuations of the amplitude, increases with the increasing nonclassicality in the system [6].

Probably, antibunching and squeezing are the most popular examples of nonclassical states and people have shown serious interest on these states since 1960s. But higher order extension of these nonclassical states are only introduced in late 1980s [7-10]. Among these higher order nonclassical effects higher

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order squeezing is studied in detail [7, 8, 11, 12] but the higher order antibunching [9] is not yet studied rigorously. As a result, still we do not have answers to certain fundamental questions like: Whether higher order antibunching and higher order squeezing appears simultaneously or not? The present work aims to provide answers to this question. In order to do so, in section 2, we have modeled the interaction of an intense laser beam with an inversion symmetric third order nonlinear medium as a quartic anharmonic oscillator. A first order operator solution of the model Hamiltonian is also used to provide time evolution of some useful operators. In section 3 and 4 we have theoretically studied the possibilities of generation of the higher order squeezed and higher order antibunched states respectively. We have shown that the generation of higher order squeezed and higher order antibunched states is possible but they may or may not appear simultaneously. We finish with some comments and concluding remarks in section 5.

## 2 The model: an intense laser beam interacts with a 3rd order nonlinear medium

An intense electromagnetic field interacting with a dielectric medium induces a macroscopic polarization  $(\overrightarrow{P})$  having a general form

$$\overrightarrow{P} = \chi_1 \overrightarrow{E} + \chi_2 \overrightarrow{E} \overrightarrow{E} + \chi_3 \overrightarrow{E} \overrightarrow{E} \overrightarrow{E} + \dots$$
 (1)

where  $\vec{E}$  is the electric field and  $\chi_i$  is the i-th order susceptibility. Corresponding electromagnetic energy density is given by

$$H_{em} = \frac{1}{8\pi} \left[ (\overrightarrow{E} + 4\pi \overrightarrow{P}) . \overrightarrow{E} + \overrightarrow{B} . \overrightarrow{B} \right]$$
 (2)

where  $\overrightarrow{B}$  is the magnetic field. Now, if we consider an inversion symmetric medium then even order susceptibilities  $(\chi_2, \chi_4 \text{ etc.})$  would vanish. Hence the leading contribution to the nonlinear polarization in an inversion symmetric medium comes through the third order susceptibility  $(\chi_3)$ . If we neglect the macroscopic magnetization (if any) then the interaction energy will be proportional to the 4-th power of the electric field. Normal mode expansion of the field ensures that the electric field operator  $E_x$  for x-th mode is proportional to  $(a_x + a_x^{\dagger})$  and the free field Hamiltonian is

$$H_0 = \sum_x \omega_x (a_x^{\dagger} a_x + \frac{1}{2}) \tag{3}$$

where we have chosen  $\hbar = 1$ .

Thus the total Hamiltonian of a physical system in which a single mode of intense electromagnetic field having unit frequency interacts with a 3rd order nonlinear non-absorbing medium is

$$H = (a^{\dagger}a + \frac{1}{2}) + \frac{\lambda}{16}(a^{\dagger} + a)^{4}$$
  
=  $\frac{X^{2}}{2} + \frac{\dot{X}^{2}}{2} + \frac{\lambda}{4}X^{4}$  (4)

with

$$X = \frac{1}{\sqrt{2}}(a^{\dagger} + a) \tag{5}$$

and

$$\dot{X} = -\frac{i}{\sqrt{2}}(a^{\dagger} - a). \tag{6}$$

The parameter  $\lambda$  is the coupling constant and is a function of  $\chi_3$ . Here we can note that the silica crystals which are used to construct optical fibers are example of third order nonlinear medium. So third order nonlinear medium described by the Hamiltonian (4) is also important from the point of view of applicability.

The above Hamiltonian (4) represents a quartic anharmonic oscillator of unit mass and unit frequency. The equation of motion corresponding to (4) is

$$\ddot{X} + X + \lambda X^3 = 0 \tag{7}$$

which can not be solved exactly. But in the interaction picture the potential  $V_I$  and the time evolution operator  $U_I$  corresponding to (4) are respectively

$$V_I(t) = \exp(ia^{\dagger}at)\lambda(a+a^{\dagger})^4 \exp(-ia^{\dagger}at) = \lambda(a\exp(-it) + a^{\dagger}\exp(it))^4$$
 (8)

and

$$U_I(t) = 1 - i \int_0^t V_I(t_1) dt_1 + (-i)^2 \int_0^t V_I(t_1) dt_1 \int_0^{t_1} V_I(t_2) dt_2 + \dots$$
 (9)

Now if we assume

$$\int_{0}^{t} V_{I}(t_{1})dt_{1} \int_{0}^{t_{1}} V_{I}(t_{2})dt_{2} \ll 1$$
(10)

then we can neglect higher order terms and (9) reduces to

$$U_I(t) = 1 - i \int_0^t V_I(t_1) dt_1 = 1 - i \int_0^t \lambda (a \exp(-it_1) + a^{\dagger} \exp(it_1))^4 dt_1$$
 (11)

Thus the first order expression for time evolution of annihilation operator is

$$a_{I}(t) = U_{I}^{\dagger}(t)a(0)U_{I}^{\dagger}(t) = a - \frac{i\lambda}{8} \left[ 6ta + 6ta^{\dagger}a^{2} + 6\exp(it)\sin ta^{\dagger 2}a - \exp(2it)\sin(2t)a^{\dagger 3} + 6\exp(it)\sin ta^{\dagger} + 2\exp(-it)\sin ta^{3} \right].$$
(12)

The derivation of this first order expression (for a more generalized Hamiltonian) is shown in detail in [13, 14]. Here we would like to note that the annihilation operator in the Heisenberg picture  $a_H$  and that in the interaction picture  $a_I$  are related by  $a_H = \exp(-it)a_I(t)$  and our solution (12) is valid only when (10) is satisfied. Physically this condition implies that the anharmonic term present in (4) provides only a small perturbation. This assumption is justified because in a third order nonlinear medium the anharmonic constant  $\lambda$ , which is a function of  $\chi_3$ , is very small (i.e  $\lambda \ll 1$ ) [14].

#### 3 Higher order squeezing

Higher order squeezing is defined in various ways. The definition which we have used in this work is by Hillery [11]. This definition is different from that of Hong and Mandel [7]. Present definition is also called amplitude squared squeezing. According to this definition of higher order squeezing, higher order quadrature variables are defined as

$$Y_1 = \frac{1}{\sqrt{2}}(a^{\dagger 2} + a^2) \tag{13}$$

and

$$Y_2 = \frac{i}{\sqrt{2}}(a^{\dagger 2} - a^2) \tag{14}$$

From the commutation relation  $[Y_{1'}Y_2] = i(4N+2)$  it is easy to conclude that a state is squeezed in  $Y_1$  variable if

$$(\triangle Y_1)^2 < \langle 2N + 1 \rangle \tag{15}$$

or if,

$$f = (\triangle Y_1)^2 - \langle 2N + 1 \rangle < 0 \tag{16}$$

A strenuous but straight forward operator algebra yields

$$\Delta Y_{1}^{2} = \langle Y_{1}^{2} \rangle - \langle Y_{1} \rangle^{2} = \begin{bmatrix} 2|\alpha|^{2} + 1 - \frac{\lambda}{4} \left[ 4|\alpha|^{2} \left( 2|\alpha|^{2} + 3 \right) \sin t \sin(2\theta - t) - 12|\alpha|^{4} t \sin(4\theta) + 3 \left( 2|\alpha|^{4} + 4|\alpha|^{2} + 1 \right) \sin^{2} 2t - 12|\alpha|^{2} \left( 2|\alpha|^{2} + 3 \right) \sin t \sin(2\theta + t) - 2|\alpha|^{4} \sin 2t \sin\left( 2(2\theta - 2t) \right) \end{bmatrix}$$

$$(17)$$

where  $\alpha = |\alpha| \exp(i\theta)$  is used. Here we would like to note that since we are working in interaction picture we have to take all the expectation values with respect to the initial coherent state  $|\alpha\rangle$  which is defined as  $a|\alpha\rangle = \alpha|\alpha\rangle$ . By taking the expectation value of  $N(t) = a^{\dagger(t)}a(t)$  we obtain

$$\langle N(t)\rangle = |\alpha|^2 - \frac{\lambda}{4} \left[ 2|\alpha|^2 \left( 2|\alpha|^2 + 3 \right) \sin t \sin(2\theta - t) - |\alpha|^4 \sin 2t \sin\left( 2(2\theta - 2t) \right) \right]. \tag{18}$$

Substituting (17) and (18) in (16) we obtain a closed form analytic expression for f as

$$f = -\frac{3\lambda}{4} \left[ -4|\alpha|^2 (2|\alpha|^2 + 3) \sin t \sin(t + 2\theta) - 4|\alpha|^4 t \sin(4\theta) + (2|\alpha|^4 + 4|\alpha|^2 + 1) \sin^2(2t) \right]$$
(19)

From (19) we can observe that f oscillates between positive and negative values depending upon the phase of the input coherent light  $\theta$  and the interaction time t. Both of these parameters can be tuned to produce higher order squeezed state and to increase the depth of noclassicality by increasing the negativity of f. If we consider  $\theta = \frac{\pi}{2}$  then (19) reduces to

$$f = -\frac{3\lambda}{4} \left[ 4|\alpha|^2 (2|\alpha|^2 + 3)\sin^2 t + (2|\alpha|^4 + 4|\alpha|^2 + 1)\sin^2(2t) \right]$$
 (20)

which is always negative and thus we always have higher order squeezing.

#### 4 Higher order antibunching of photons

As we have already mentioned the higher order antibunching [9] is not yet studied rigorously. Using the negativity of P function [1], Lee introduced the criterion for HOA as

$$R(l,m) = \frac{\left\langle N_x^{(l+1)} \right\rangle \left\langle N_x^{(m-1)} \right\rangle}{\left\langle N_x^{(l)} \right\rangle \left\langle N_x^{(m)} \right\rangle} - 1 < 0, \tag{21}$$

where N is the usual number operator,  $N^{(i)} = N(N-1)...(N-i+1)$  is the ith factorial moment of number operator,  $\langle \rangle$  denotes the quantum average, l and m are integers satisfying the conditions  $l \leq m \leq 1$  and the subscript x denotes a particular mode. Ba An [15] choose m=1 and reduced the criterion of lth order antibunching to

$$A_{x,l} = \frac{\left\langle N_x^{(l+1)} \right\rangle}{\left\langle N_x^{(l)} \right\rangle \left\langle N_x \right\rangle} - 1 < 0 \tag{22}$$

or,

$$\left\langle N_{x}^{(l+1)} \right\rangle < \left\langle N_{x}^{(l)} \right\rangle \left\langle N_{x} \right\rangle.$$
 (23)

Physically, a state which is antibunched in lth order has to be antibunched in (l-1)th order. Therefore, we can further simplify (23) as

$$\left\langle N_x^{(l+1)} \right\rangle < \left\langle N_x^{(l)} \right\rangle \left\langle N_x \right\rangle < \left\langle N_x^{(l-1)} \right\rangle \left\langle N_x \right\rangle^2 < \left\langle N_x^{(l-2)} \right\rangle \left\langle N_x \right\rangle^3 < \dots < \left\langle N_x \right\rangle^{l+1} \tag{24}$$

and obtain the condition for l-th order antibunching as

$$d(l) = \left\langle N_x^{(l+1)} \right\rangle - \left\langle N_x \right\rangle^{l+1} < 0. \tag{25}$$

This simplified criterion (25) coincides exactly with the physical criterion of HOA introduced by Pathak and Garica [16].

By using (12) and the condition for higher order antibunching (25), it is easy to derive that for a third order non-linear medium having inversion symmetry, we have

$$d(1) = \frac{3\lambda |\alpha|^2}{4} \left[ 2\left(2|\alpha|^2 + 1\right) \sin(t - 2\theta) \sin(t) + |\alpha|^2 \sin(2(t - 2\theta)) \sin(2t) \right],$$
(26)

$$d(2) = \frac{3\lambda |\alpha|^4}{2} \left[ \sin(t - 2\theta) \sin(t) + \sin(2(t - 2\theta)) \sin(2t) \right]$$
 (27)

and

$$d(3) = \frac{3\lambda |\alpha|^4}{4} \sin(2(t - 2\theta)) \sin(2t). \tag{28}$$

Antibunching of fourth or higher order can not be observed with a first order solution of the model Hamiltonian. In order to study the possibilities of their occurrence we have to use higher order operator solutions of (7).

Equations (26-28) coincides exactly with our recent result [16] which was reported as a special case of a generalized Hamiltonian. Here we can observe that if we choose interaction time  $t=2\theta$  then d=0. Therefore, we can observe higher order coherence. But for  $\theta=0$  or  $\theta=n\pi$ , (i.e. when input is real) d is a sum of square terms only. So d is always positive and we have higher order bunching of photons. For other values of phase  $(\theta)$  of input radiation field, value of d oscillates from positive to negative, so we can observe higher order bunching, anti-bunching or coherence in the output depending upon the interaction time t.

### 5 Summary and concluding remarks

From (20) we know that for  $\theta = \frac{\pi}{2}$  we always obtain higher order squeezed state. But for  $\theta = \frac{\pi}{2}$ , (27) and (28) reduces to

$$d(2) = \frac{3\lambda |\alpha|^4}{2} \left[ -\sin^2(t) + \sin^2(2t) \right]$$
 (29)

and

$$d(3) = \frac{3\lambda |\alpha|^4}{4} \sin^2(2t). \tag{30}$$

respectively. Here we can see that d(3) is always positive for this particular choice of  $\theta$ , and therefore, third order antibunching will not appear simultaneously with the amplitude squared squeezing. On the other hand d(2) oscillates between positive (higher order bunching) and negative (higher order antibunching) values. Thus we can conclude that neither second and third order antibunching appears simultaneously nor the higher order squeezing appears simultaneously with the higher order antibunching. Alternatively, we can state that, in general higher order nonclassical effects may appear separately. The possibility of their appearance may be tuned by tuning the phase  $(\theta)$  of the input coherent state and interaction time (t).

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